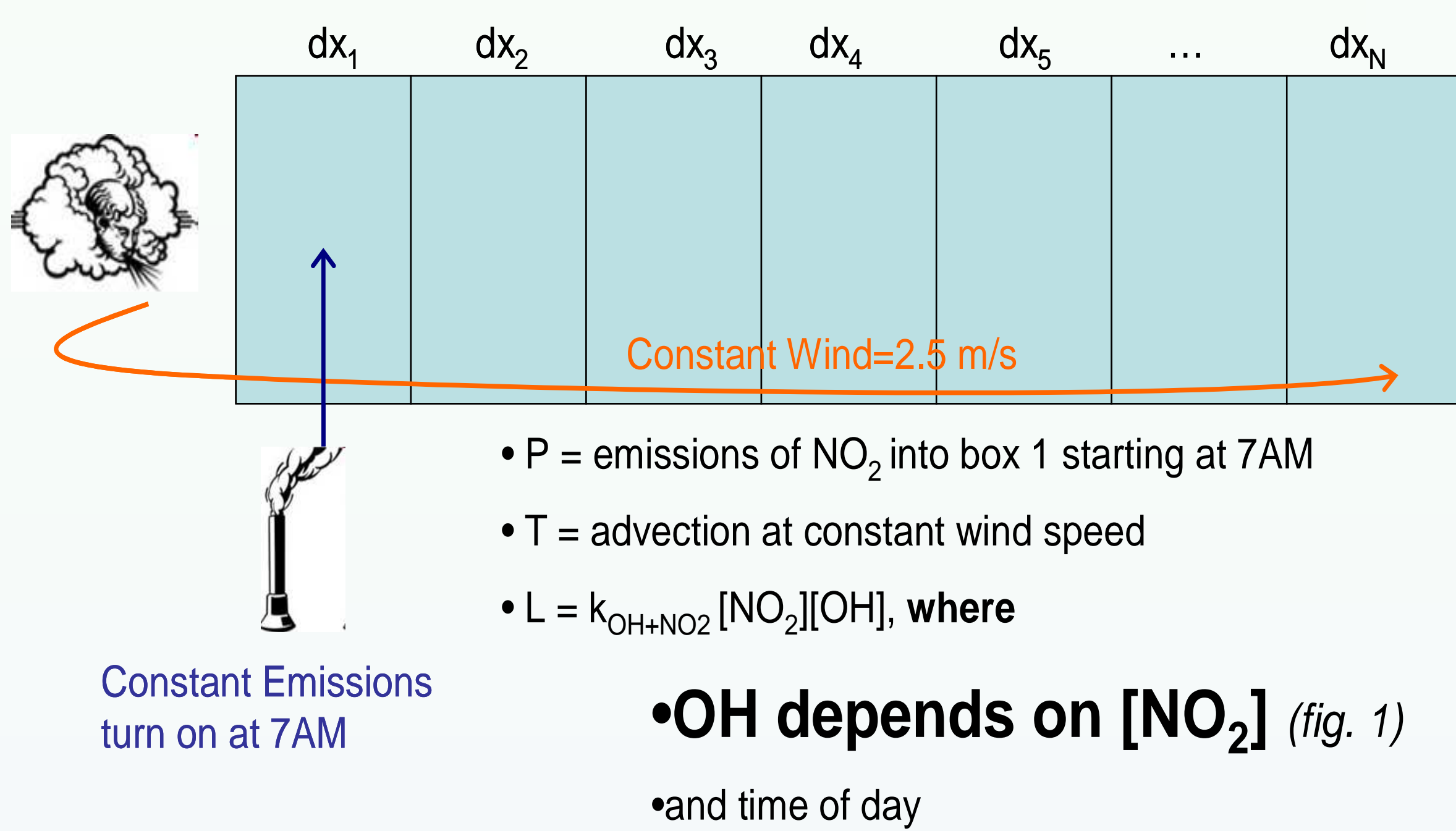# The spatial resolution needed from models and satellite-based measurements to accurately predict NO$_x$ emissions

Lukas C Valin, Ashley R Russell, Pascal M Tay, Simon A Schmutz, Ronald C Cohen

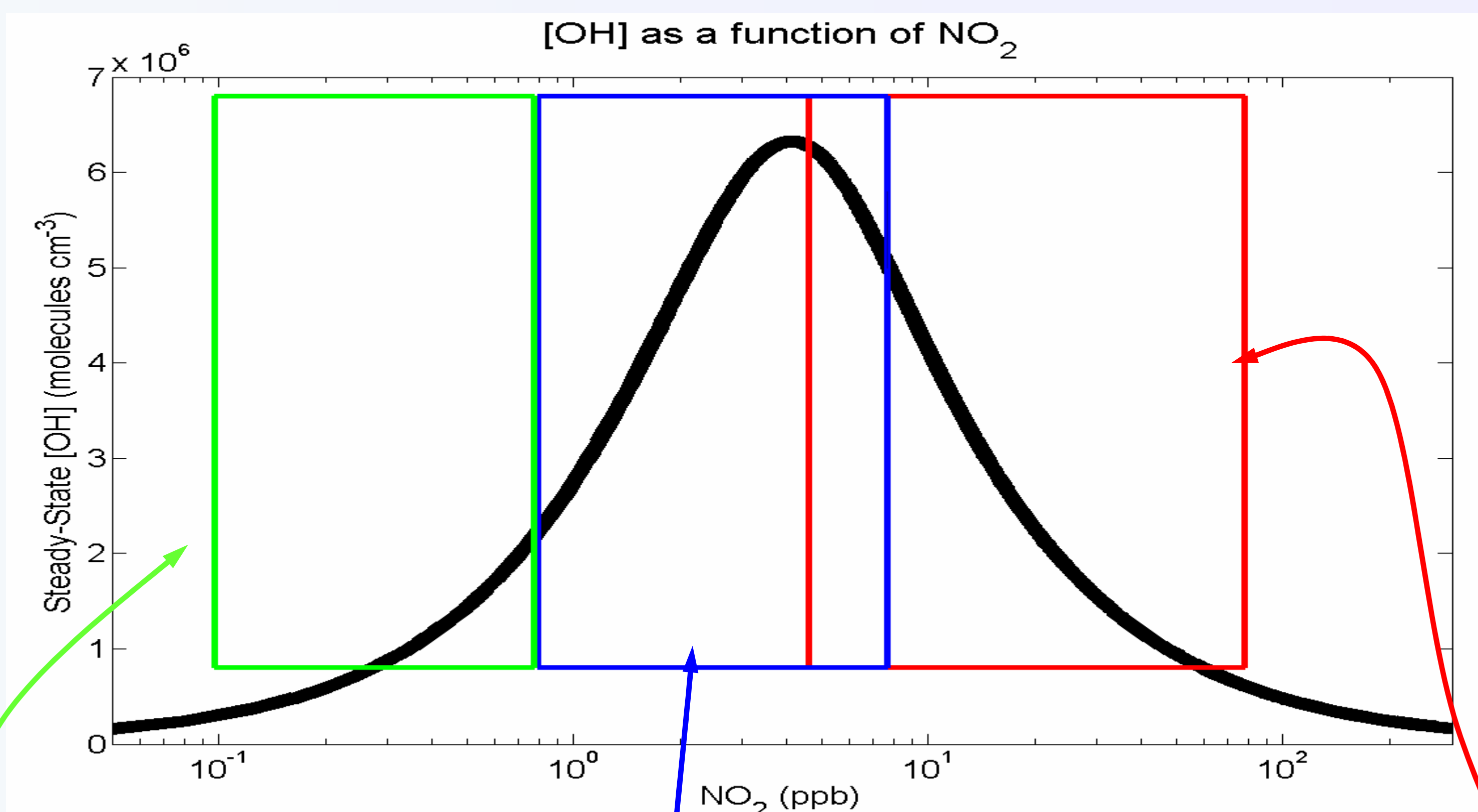College of Chemistry, University of California, Berkeley, CA 94720

Introduction

The tropospheric NO$_2$ column, a quantity accessible from space-based measurements, varies in space and time due to emissions, transport and also the non-linear response of OH to NO$_x$ concentration. We show that in a simple plume model describing the near field of cities there are steep nonlinearities in [NO$_2$]/unit emissions because of this OH feedback. At spatial scales of 10-50 km such effects are observable in 3-dimensional chemical transport models. We derive the model resolution necessary to infer emissions from space-based observations to 25% accuracy. The OMI detection limit of 1x10^{15} molecules cm^{-2} corresponds to 0.4 ppb surface concentration in a well mixed 1km boundary layer at sea level.
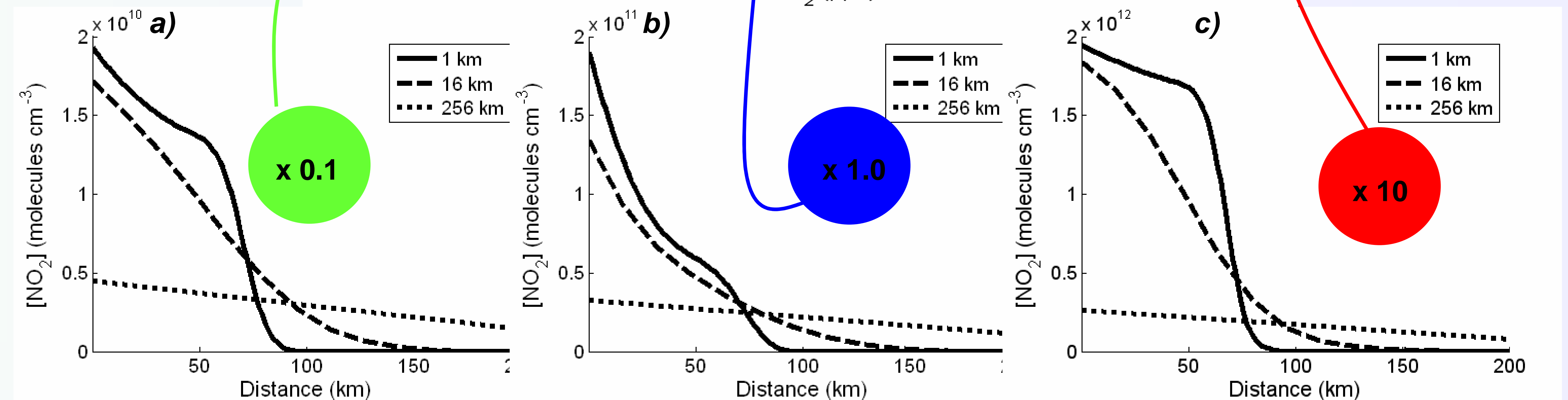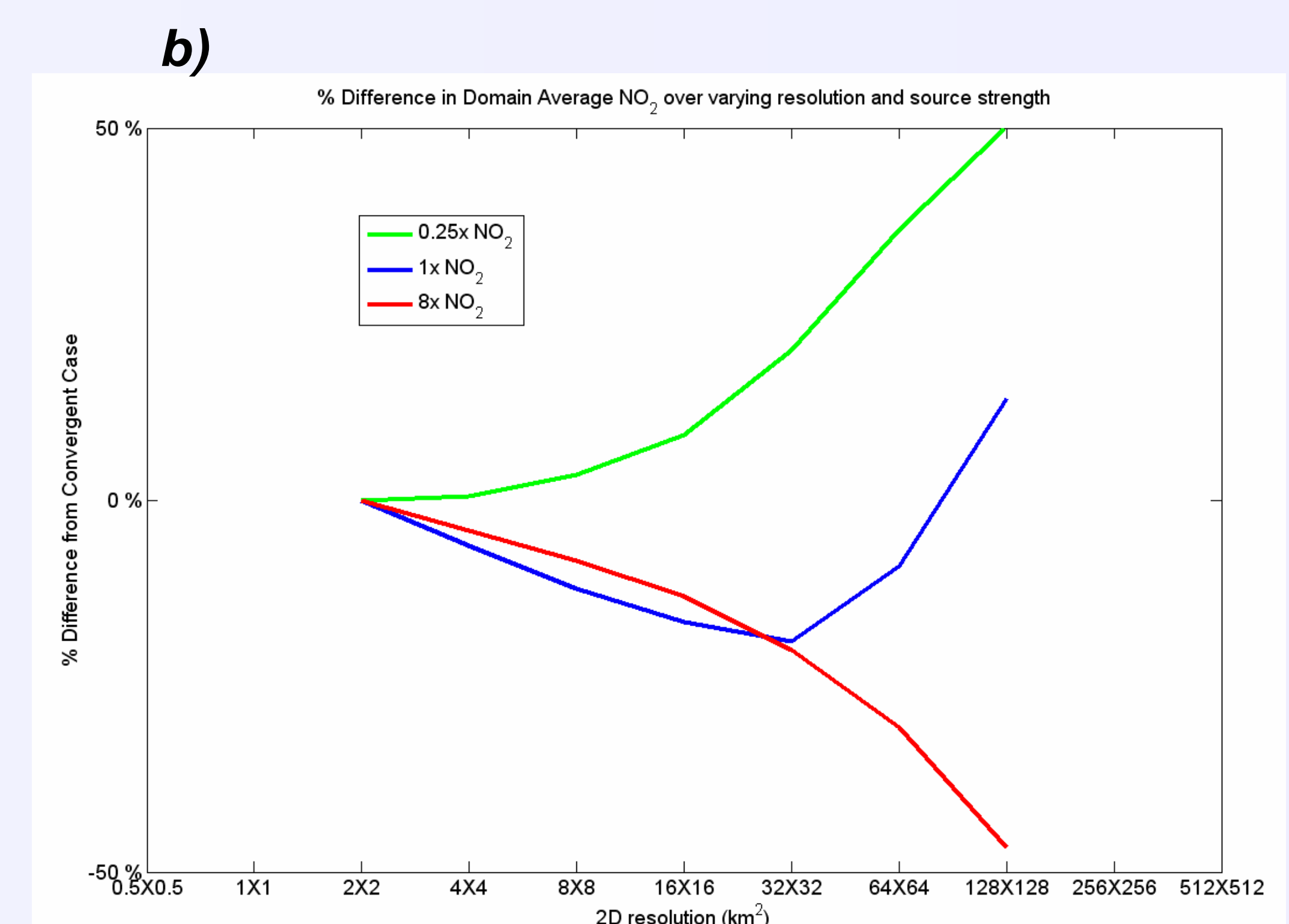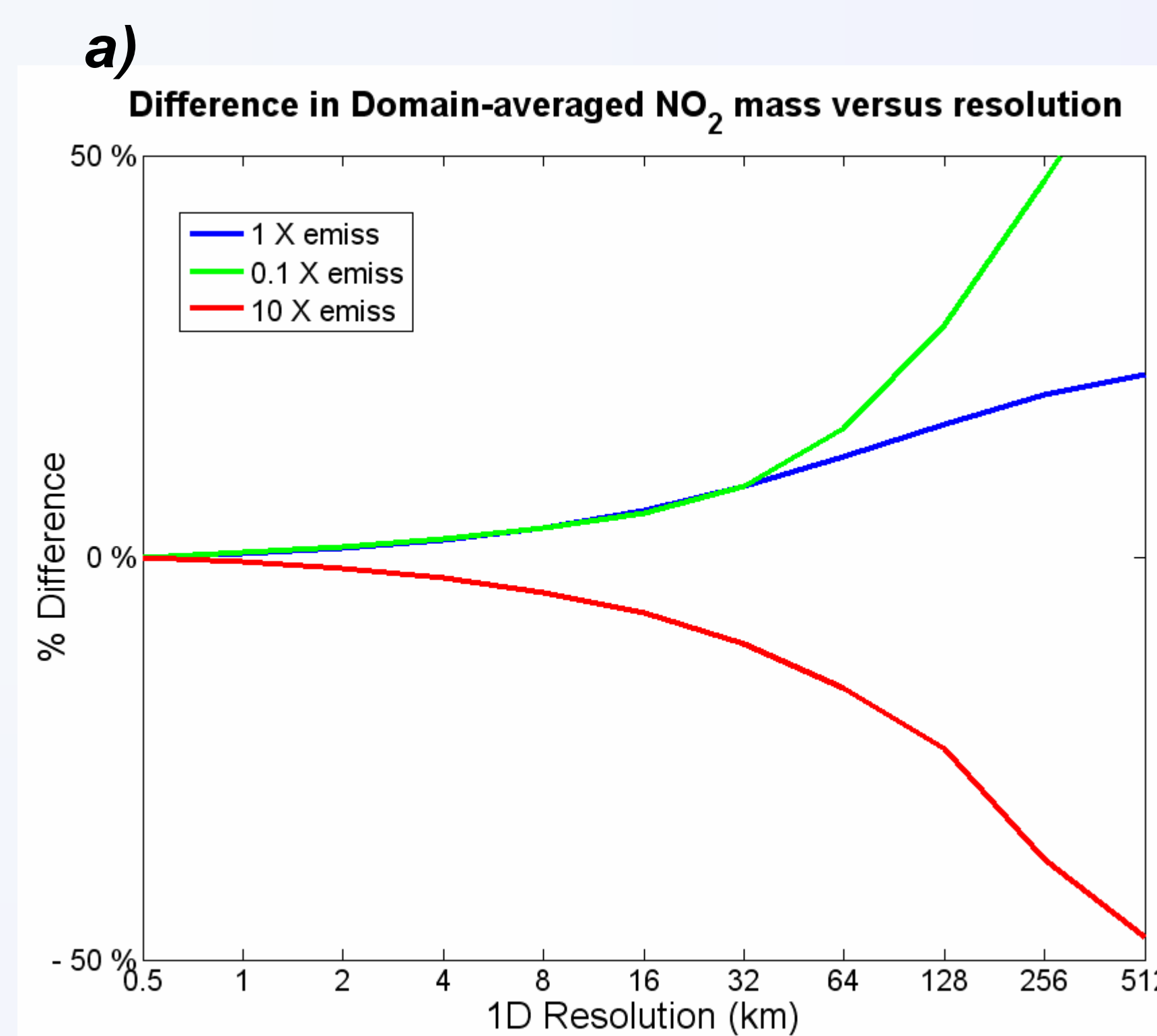
Figure 1 Analytical solution for steady-state [OH] as a function of NO$_2$[1]. The solution assumes constant pHO$_x$, k$_{OH+VOC}$[VOC], and [NO$_2$]/[NO]. Boxes indicate NO$_2$ concentrations where OH is linear in NO$_2$ (green), relatively flat (blue), and inversely proportional to NO$_2$ (red).

1D Advection and Nonlinear [OH] Response

Constant Emissions turn on at 7AM

Constant Wind=2.5 m/s

- P = emissions of NO$_2$ into box 1 starting at 7AM
- T = advection at constant wind speed
- L = k$_{OH+NO2}$ [NO$_2$][OH], **where**

•OH depends on [NO$_2$] *(fig. 1)*

•and time of day

•How does Near-Field and domain-averaged [NO$_2$] vary with Resolution?

Exploring Resolution Effects in a 3D CTM

•Horizontal and vertical mixing should increase diffusion at plume edges (2D vs 1D) and differing vertical mixing (3D)
•Terrain features, and thus transport, will vary with resolution.
•Predict large near-field disagreements between different resolutions.

Figure. 2 Ground-level [NO$_2$] (molecules cm^{-3}) versus distance (km) at 1 PM of 1D advection model for resolutions of 1, 16, and 256 km for **a)** 0.1x **b)** 1.0x **c)** and 10x emissions cases. Steep nonlinearities in the near-field decay are evident. The apparent [NO$_2$] does not vary linearly with either emissions or model resolution

Figure 3 a) Percent difference domain-averaged [NO$_2$] versus model resolution for three emissions cases at 1PM in a 1D advection model. **b)** Percent-difference domain-averaged [NO$_2$] versus resolution for three emissions cases at 1PM in a 2D advection model. The domain-averaged [NO$_2$] is more sensitive to resolution in the 2D case as diffusion plays a larger role. The local minimum in the 1x emission case is related to the point where apparent dilution created by the lower resolution causes more grid cells to have concentrations in the low NO$_2$ regime *(fig 1 green box)*.

Figure 4 a) WRF-CHEM 4x4 km^2 predicted LOG$_{10}$([NO2] (molecules cm^{-2})) averaged at 1PM for July 4-9, 2005. ±25% deviation from 4^2 km^2 predictions are shown for **b)** 12^2, **c)** 24^2 **d)** 48^2 and **e)** 96^2 km^2 predictions (black → 4km case higher, gray → 4 km case lower). Large disagreements occur in regions of transition between OH response regimes to varying NO$_2$. The deviation can be attributed in part to the near-field response of [OH] to NO$_2$ as seen in the 1D and 2D case explored above. However, some spatial differences can be attributed to the different transport between cases (e.g. Difference in LA outflow between 12 and 4 km model runs).
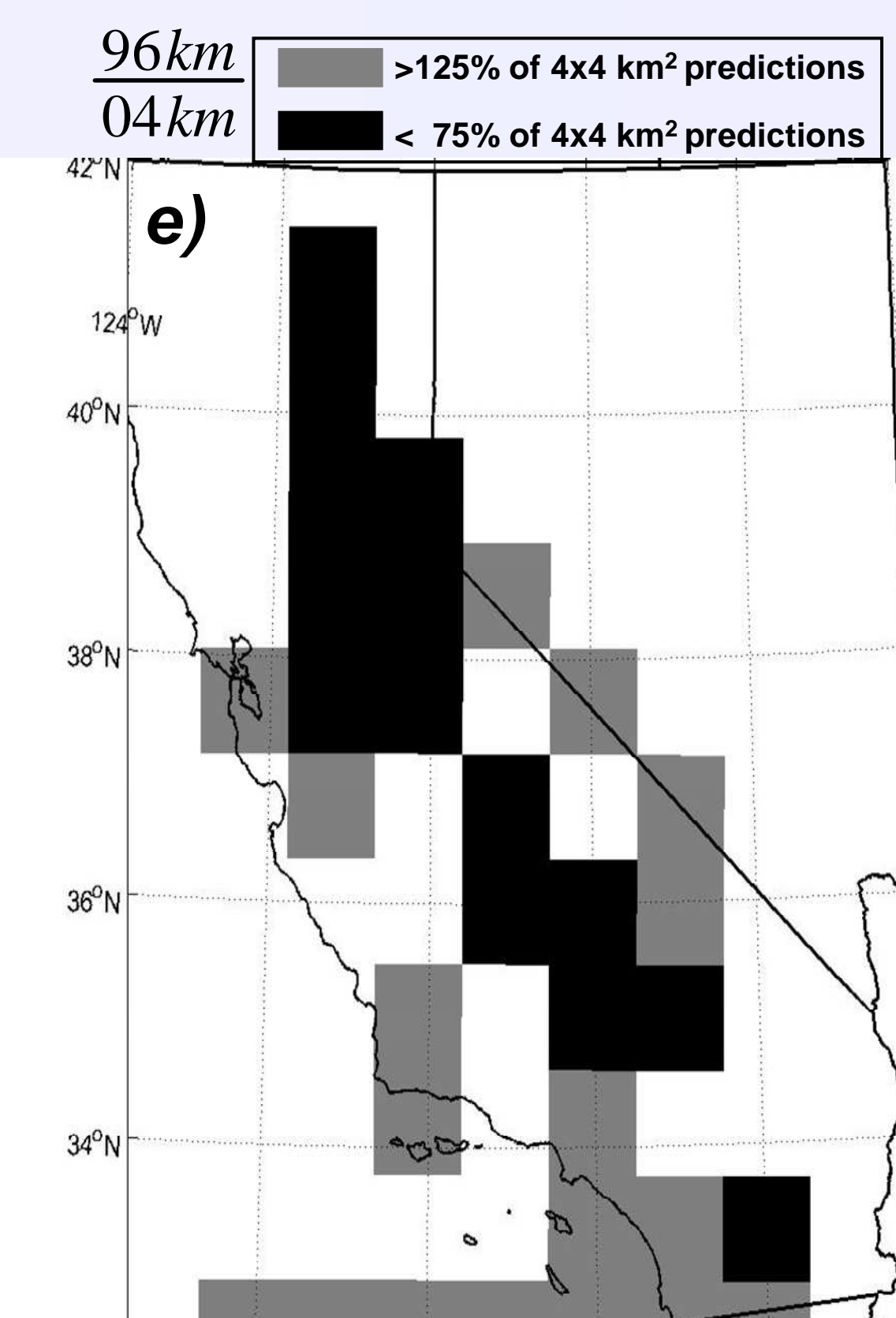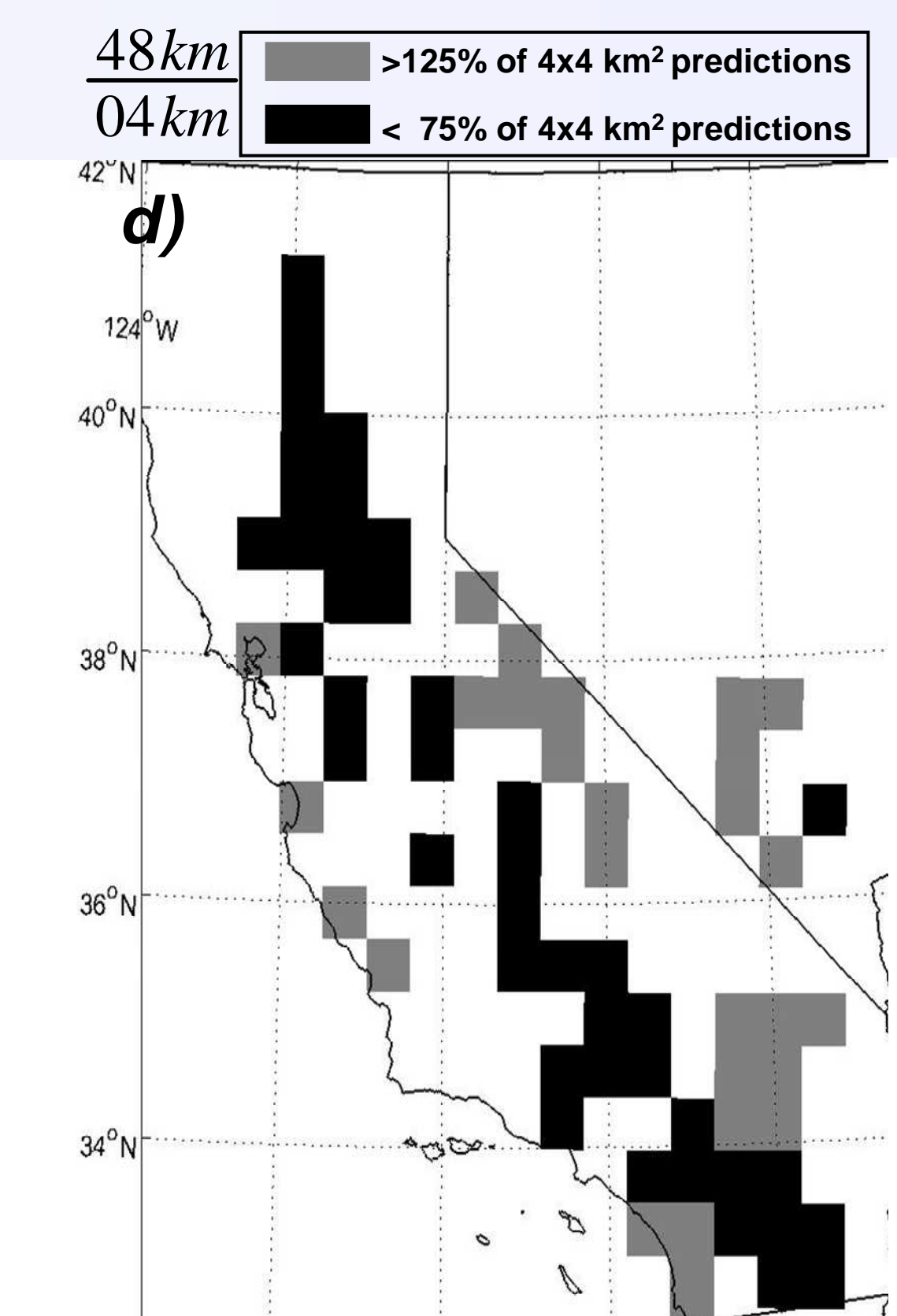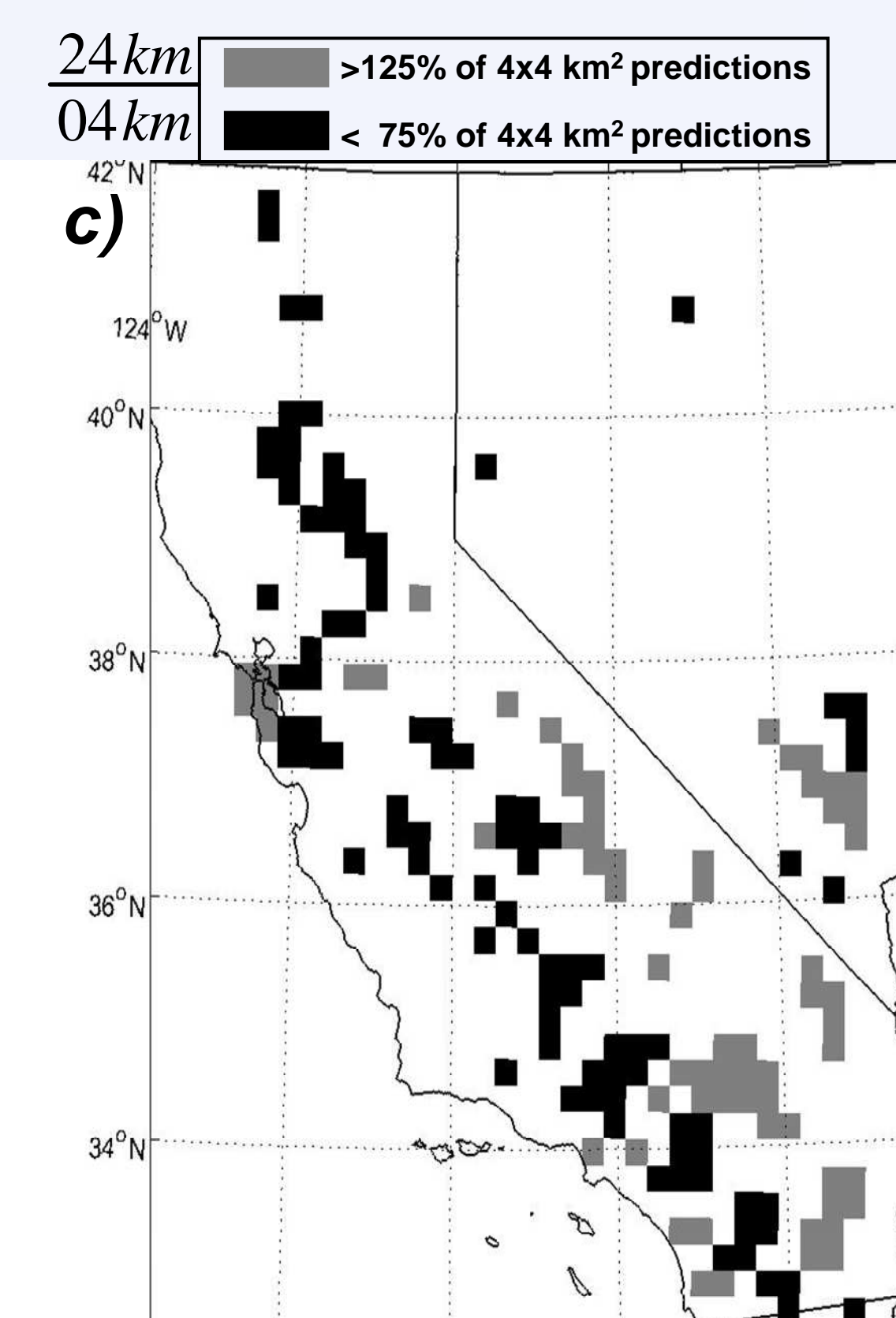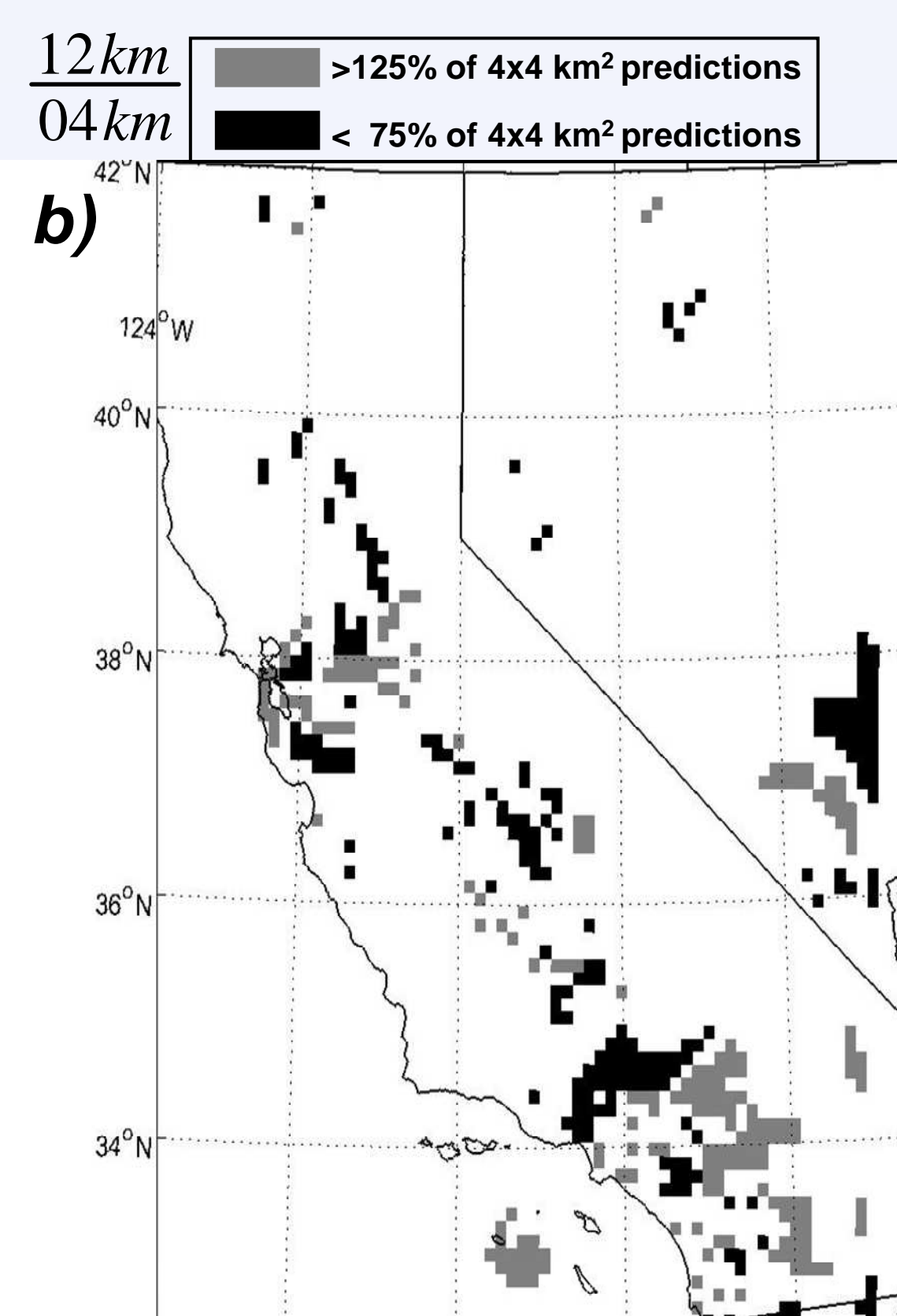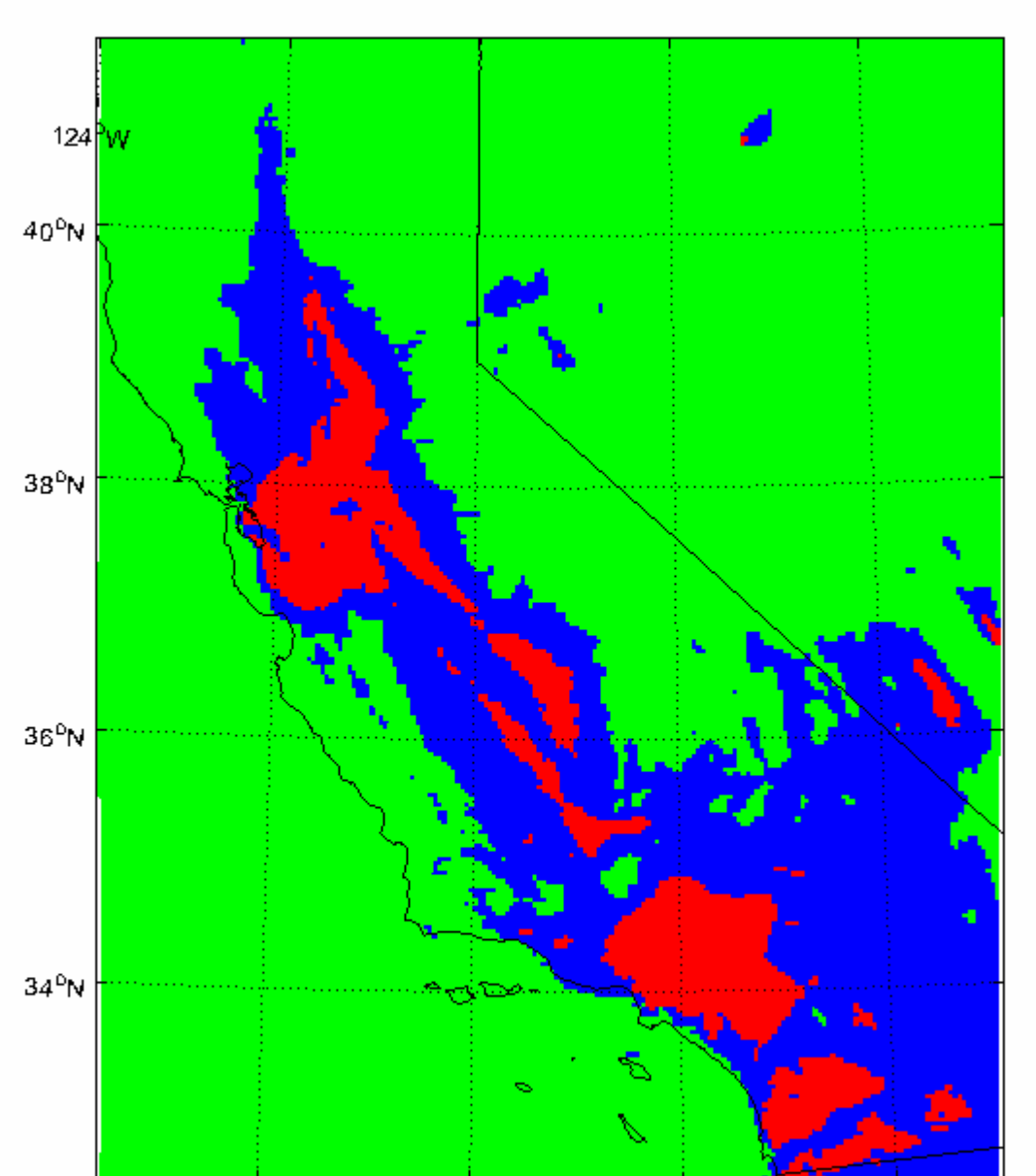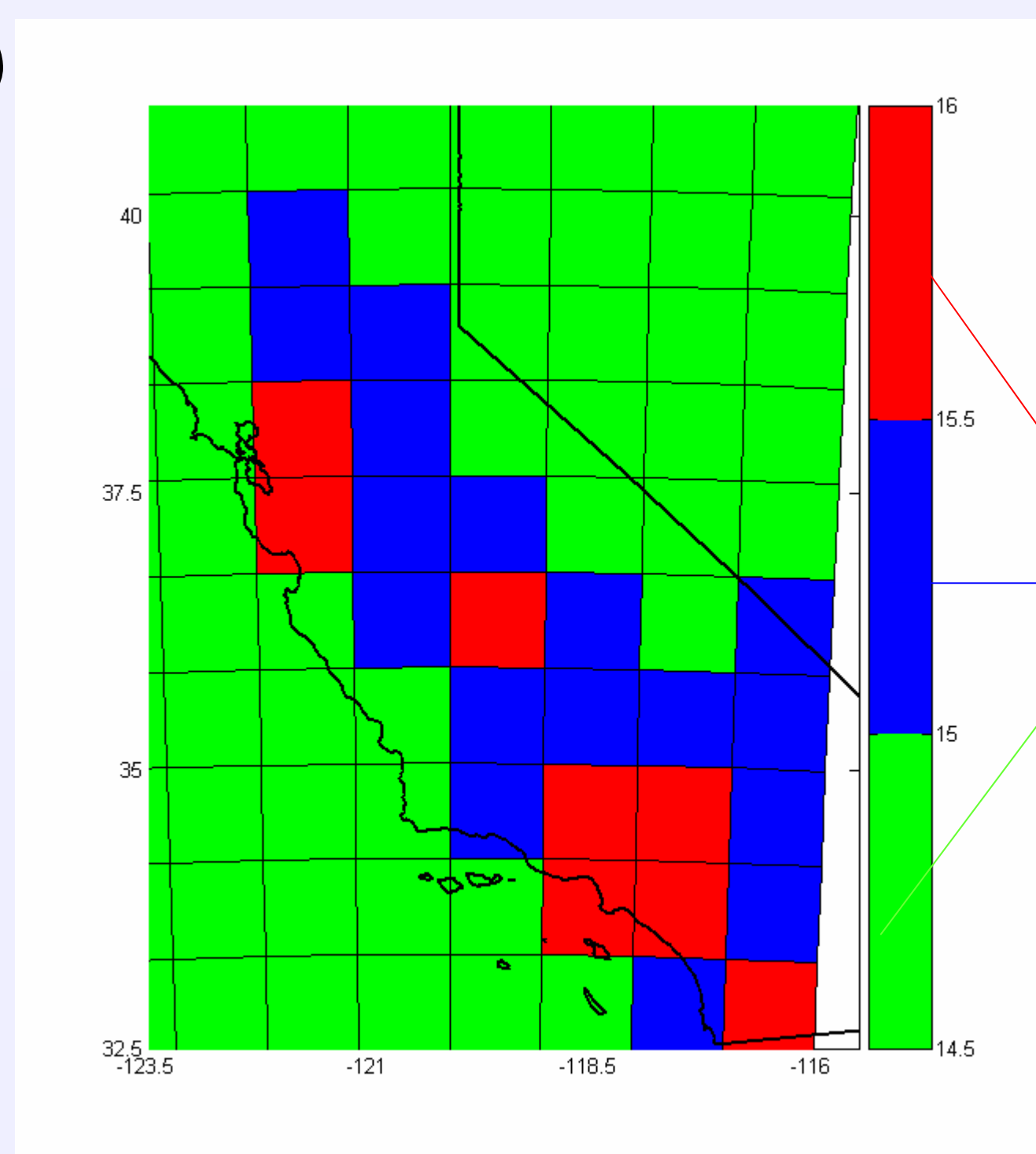
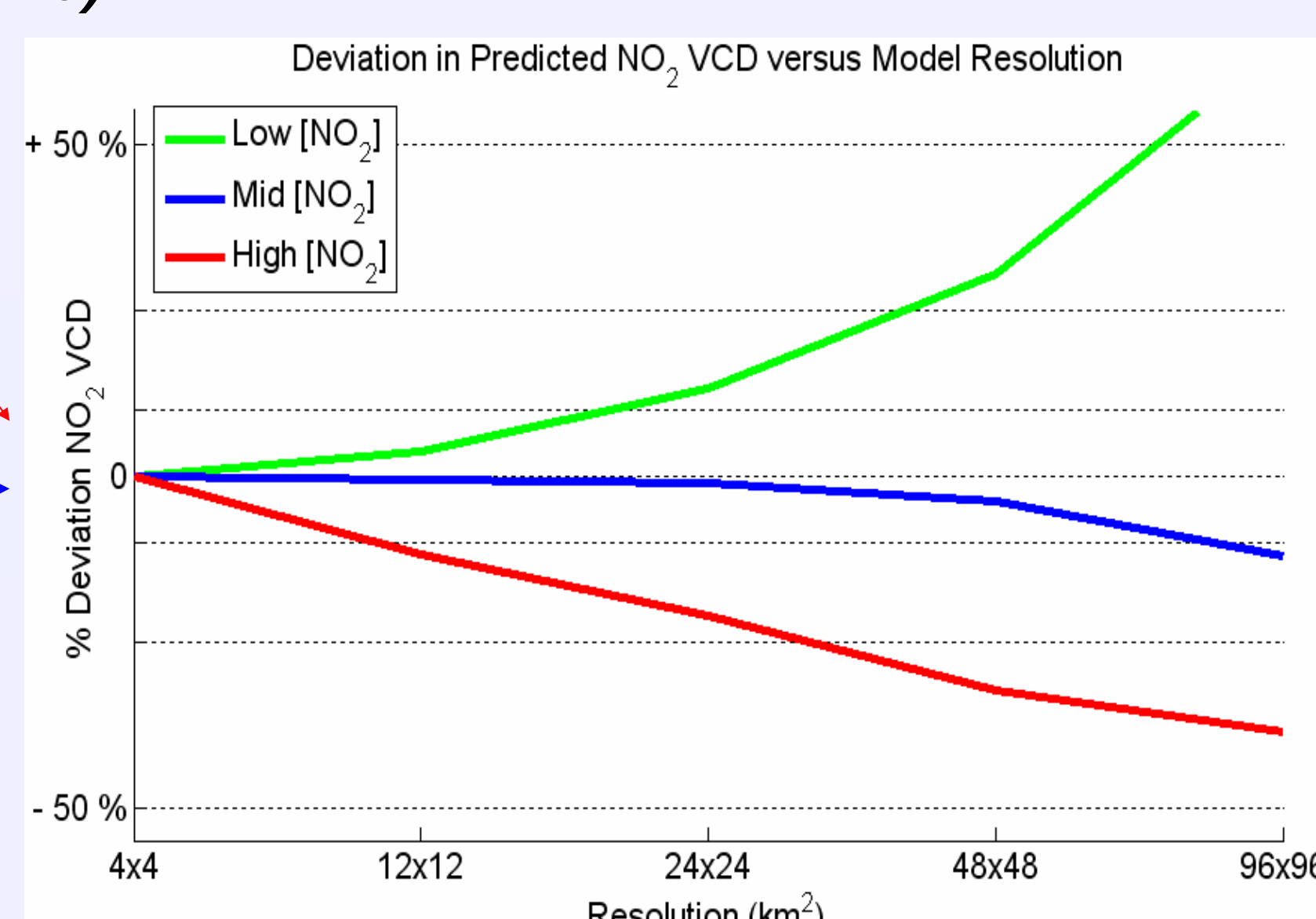**Figure 5 a)** The 4 km model run averaged to 96 km. The color scale is log$_{10}$ NO$_2$ column density (<10^{15}, 10^{15}< **x** < 10$^{15.5}$, >10$^{15.5}$,). **b)** The divergence in the average values of bins versus model resolution.

Conclusions

These results imply that **a)** large scale (1°x1°) inverse models cannot accurately (±25%) retrieve emissions, **b)** even 12 km resolution is insufficient to for model convergence of NO$_2$ columns to ~ 10%, and **c)** that OMI observations are at the threshold of having too low spatial resolution to accurately constrain emissions

Works Cited

1. Murphy, J. G., Day, D. A., Cleary, P. A., Wooldridge, P. J., Millet, D. B., Goldstein, A. H., and Cohen, R. C. *Atmos. Chem. Phys.*, 7, 5327-5339, 2007.

Acknowledgements